Sustainability in Carbon Capture and Utilization. Biodiesel from Microalgae

E. N. D'Addario

Abstract Biodiesel from microalgae is one of the most promising options of carbon capture and utilization (CCU) because of the direct utilization of solar radiation and the higher biomass and lipid areal productivity in comparison with marine or terrestrial plants. LCA studies on this subject are rather difficult to compare because of inhomogeneous assumptions for oil extraction (the critical step), fertilizer consumptions, use of residual biomass, use of primary energy needed for electricity, use of different fossil fuel for process heat supply, availability of waste heat from fossil or biomass power stations, and climate of the cultivation areas. Nevertheless, GHG savings appear in line with the current EU standards for automotive fuel only under the most favourable conditions (biomass productivity at 25 gm^{-2} day^{-1}, lipid content at 40–45 %, water and nutrient recycle, favourable climate conditions, use of low energy wet oil extraction processes still to be checked on large scale).

Keywords Microalgae · Sustainability · CCU · LCA · Biodiesel from microalgae

1 Introduction

Many options for Carbon Capture and Utilization (CCU) are currently or potentially available. Carbon dioxide has in fact already its own market as blanketing agent, fire extinguisher, drying ice, refrigerating fluid, aerosol propellant, respiratory stimulant after addition to medical O_2, shielding gas in welding, carbonation

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of beverages, extracting agent in supercritical extraction of caffeine, flavour and flagrancies, etc.

Numerous routes for conversion of carbon dioxide in bulk chemicals or intermediates such as methane, carbon monoxide, methanol and urea which can be used to get liquid fuels, fertilizers and polycarbonate plastics are also possible. Moreover, carbon dioxide can be fed in greenhouses for algae cultivation which represents one of the best photosynthetic processes capable to generate high-quality food (protein) or food supplement (omega 3 polyunsaturated fatty acids, carotenoids, vitamins, etc.) and/or biofuel (essentially lipids-derived diesel substitutes and carbohydrates-derived gasoline substitutes). Additionally, carbon dioxide can also be reacted with many purified silicon oxide based minerals such as olivine, serpentine, wallostonite or similar solid residuals from waste incineration, to get calcium or magnesium carbonates usable as building materials. Finally, carbon monoxide can be injected in partially depleted petroleum reservoir to enhance oil recovery (EOR).

These two last routes remain the most promising options for carbon dioxide utilization. In fact, one of the most interesting literature source [1] estimates that if 10 % of the total annual building material demand were replaced by products from carbonation of minerals and waste, the greenhouse gas reduction would be around 1.6 Gt CO_2 equivalent per year and enhanced oil recovery could give approximately the same contribution (1.4 Gt CO_2 equivalent per year) [1]. While, in hypothesis of replacing 5 % of current transport fuel with second and third generation biofuel having 50 % less CO_2 emission in comparison with fossil fuel, the estimated reduction of CO_2 emission would reach 0.4 Gt per year [1]. A lower contribution is estimated to be given by the production of chemical feedstock (0.3 Gt CO_2 equivalents per year) [1].

However, any of the above mentioned new routes of CO_2 utilization suffer for the rather scarce CO_2 reactivity and for water, hydrogen and energy requirements. This, with the potential partial exception of mineral and waste carbonation which are based on exothermic reaction, but needs rather energy intensive procedures for the purification of the reactant [2]. Therefore, in the near term, possible revenues from CO_2 market are still not enough to counterbalance drawbacks due to requirements of non-fossil energy sources such as nuclear or renewables (solar, wind, geothermal, tidal, hydro, etc.), with the exception of favourable enhanced oil recovery cases.

In the previous depicted panorama, one of the most promising options of CO_2 utilization can be identified in photosynthetic processes for microalgae cultivation. Microalgae in fact, contrarily to macroalgae (seaweeds), terrestrial and aquatic plants which grow on atmospheric CO_2, may be advantageously cultivated in systems such as open ponds or photobioreactors where captured carbon dioxide is bubbled [3, 4]. In this way, microalgae can reach photosynthetic efficiencies and biomass productivity very high in comparison with other cultivated biomass, as shown in Table 1 [5].

Table 1 High productivity biomass [5]

Biomass community	Location	Yield (t d.w. ha^{-1} y^{-1})	Photosynthetic efficiency (%)
Hybrid poplar	Minnesota	8–11	0.3–0.4
Water hyacinth	Mississippi	11–33 (>150)	0.3–0.9
Switch grass	Texas	8–20	0.2–0.6
Sweet sorghum	Texas-California	22–47	0.6–1.0
Coniferous forest	England	34	1.8
Maize	Israel	34	0.8
Tree plantation	Congo	36	1.0
Tropical forest	West Indies	60	1.6
Algae	Different locations	70	2–2.5
Sugar cane	Hawaii-Java	64–87	1.8–2.6
Napier grass	Hawaii, Puerto Rico	85–106	2.2–2.8

Furthermore, microalgae have good carbohydrate content (10–60 % depending on the species, as reported in Ref. [4]) and lipid content, typically in the range of 10–30 % which under specific cultivation conditions can reach very high values (45 %) [6]. For this reasons, microalgae are one of the most studied biomass as a source of second/third generation biofuel, particularly lipid-derived automotive diesel [7, 8]. However, many research efforts are still needed to circumvent technical and economical bottlenecks of this technology [9, 10], and accurate studies are necessary to assess its environmental sustainability.

Life cycle analysis (LCA) appears one of the most suitable tools usable at this last purpose. This methodology in fact, relies on well-established codes (ISO Standards 1440-1444 2006), on recognised databanks (Simapro, Ecoinvent, Bounstead) and allows the calculation of a number of parameters (impact categories) capable to bring into account the environmental impact at different level: local (toxic emissions, noise, electromagnetic pollution), regional (acid rain, visual pollution, water eutrophication, soil and groundwater contamination) and global (greenhouse effect, depletion of not renewable resources, photochemical oxidant formation, land use change, depletion of the ozone layer).

In this context, the results of a number of LCA case studies published in the recent literature on the production of diesel substitutes from microalgae are reviewed and compared, in order to identify critical aspects of current research and possible applicative scenarios.

2 General Aspects on LCA for Microalgae Production

As mentioned before, rigorous LCA studies are carried out according to very well-established criteria defined in the ISO standards 14040 and 14044. Accordingly, a selected functional value (functional unit) of the product whose impact has to be assessed is defined, and the methodology is implemented through four phases consisting of:

- goal and scope definition,
- life cycle emissions inventory and analysis,
- impact assessment,
- results interpretation.

Usually, phase two is the most challenging and relies upon a deep knowledge of the technology under examination, a proper definition of system boundaries, a detailed energy and material balance and an accurate emissions calculation. In this view, the higher the scientific and technical knowledge of the production process, the higher the LCA accuracy of the related product. In the case of LCA related to microalgae production for energy purposes, goal changed through the time, technologies have been only partially tested at laboratory pilot scale, and the development of the complete know how still require intensive research efforts. For instance, microalgae were initially considered for cultivation nearby natural gas power station, and the obtained biomass was submitted to anaerobic digestion to get methane in order to reduce natural gas consumptions [11]. After microalgae have been considered for cultivations nearby coal power plant to get biomass to be used in co-firing units and, the environmental impact of this potential industrial application has been rather accurately examined in a detailed LCA study [12].

Only in last year's microalgae have been proposed for lipid-derived automotive fuel, particularly: biodiesel and high-quality diesel (green diesel). The first product is obtained by converting only the triglycerides via trans-esterification processes, and the second by submitting the whole wet algal biomass to different high pressure thermal treatments (torrefaction, pyrolisis, hydrothermal) to get a raw so-called bio-oil which has to be extensively refined and hydrogenated to get green diesel. LCA studies published in the current literature refers principally to biodiesel and are rather inhomogeneous because of the assumptions made for: (1) microalgae cultivation (algae species, concentration in the cultures, biomass productivity, lipid content, open pond or photobioreactors cultivation systems and related engineering, construction materials, operations, etc.), (2) systems for microalgae harvesting and dewatering (centrifuges, gravity settlers under different pH, use of synthetic flocculants, belt or filter press, etc.), (3) hypotheses used to calculate nutrient consumption and accordingly water recycle options and (4) processes used for lipid extraction.

In the following, assumptions and results published in one of the most interesting LCA study on biodiesel production from microalgae [13] are summarized and compared to more recent works.

3 Basic Aspects of Biodiesel Production from Microalgae

3.1 *Microalgae Cultivation and Harvesting*

A typical block diagram for biodiesel production from microalgae has been proposed by Lardon et al. [13] (Fig. 1). According to this diagram and data reported in Ref. [13], microalgae are supposed to be cultivated in 100 ha, 10 m large, 100 m long open raceway ponds, operated at 0.3 m water level and mixed with electric paddlewheel at 0.25 m/s. Ponds are supposed to be realized in concrete, and a CO_2 contained in a flue gas stream coming from a 50 MW coal power station dehydrated, and compressed was envisaged to be used for photosynthesis. Total electric energy consumption for this operation was estimated to be 22.2 Wh/kg CO_2. Evaporation was assumed to be 300 mm per annum, and water was envisaged to be totally removed each two months from ponds in order to manage contamination and prevent accumulation of toxic and inhibitory molecules. Excess water is treated in a typical active sludge wastewater treatment plant. Ponds were managed under typical nitrogen supply condition (normal culture) and under nitrogen starvation (Low N culture) which is currently proposed in the current literature to increase lipid content in the produced biomass [6]. Accordingly, residence times for a typical microalgae (*Chlorella vulgaris*) were set at 0.99 day for normal culture and 0.77 day for Low N

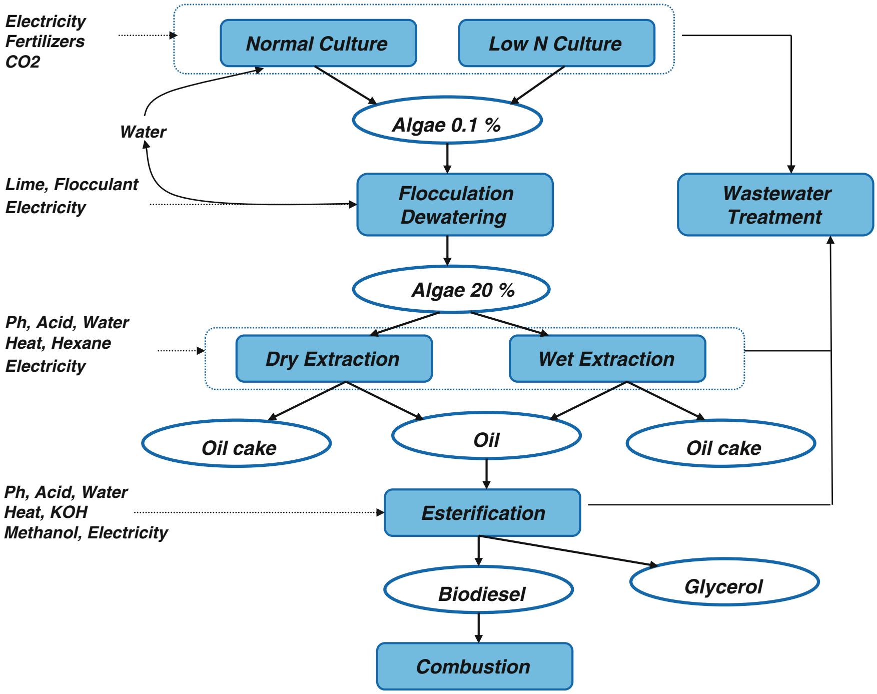

Fig. 1 Typical block diagram for biodiesel production from microalgae [13]

cultures. Under this conditions, biomass concentration in both cultures was assumed to be 1 g dry weight per litre, and productivity 24.75 and 19.75 gm^{-2} day^{-1}, respectively, for normal cultures and Low N cultures. Lipid content in normal and Low N cultures was set at 17.5 and 38.5 %.

Nitrogen consumptions were calculated according to the elemental formula (CHNO) of protein, carbohydrates and lipid assumed to be contained in the algal biomass, and minerals (phosphorous, magnesium, potassium and sulphur) were proportionally derived (see Ref. [13] for details).

Microalgae harvesting was supposed to be carried out by pH adjustment with lime (pH 11, 300 g lime/m^3) and flocculants addition (0.5 g/m^3) in typical settling units; then, the biomass concentration was increased up to 20 % dry weight basis in dewatering units. Water from harvesting operations was recycled to the cultures, and water from dewatering was purged and treated in a biological wastewater treatment plant. The total water consumption due to evaporation, purge, bimonthly total water change and residual water in the biomass was assumed to be 4 L per kg dry weight algae. Details on the process above are reported in Ref. [13].

According to experimental data obtained in long test runs carried out in a large scale pilot facility, previous assumptions, both for growth and biomass harvesting, appear realistic, a part those related to low nitrogen cultures, which are rather difficult to be maintained in long term experiments [14–16].

3.2 Oil Extraction and Lipid Trans-Esterification

A number of technologies, basically derivable from oil extraction from dry (rapeseed, soybean, peanuts, etc.) and wet oily seeds (palm and olive oil), are potentially available for lipid extraction from microalgae. Nevertheless, due to the nature of cells, these technologies do not appear usable for wet microalgae paste. Therefore, considering the high energy demand of algae drying also after dewatering, numerous research efforts are still in progress on the subject [7–10], and new technologies are being developed [17–19], but none of them have been tested at industrial scale. In this context, the two options for lipid extraction adopted by Lardon et al. [13] shown in Fig. 1 and briefly summarized in the following appear reasonable for LCA purposes. In fact, the first option is based on the use of solvent (hexane) extraction from dry biomass which is technically feasible but energy intensive and can be considered the worst case. The second assumes the utilization of wet biomass and envisages process data which have to be confirmed. Thus, it can be considered as a long term hypothesis.

Drying was supposed to be carried out in a belt dryer, a typical equipment used for sludge drying in wastewater treatment plants. The dry matter content was anticipated to be increased from 20 to 90 % (the typical value of vegetable seeds) by consuming 400 Wh/kg dry matter electricity and 13.8 MJ/kg dry matter natural gas heat.

In the oil extraction process with dry biomass, hexane loss was set at 2 g/kg dry algae and heat and electricity consumptions were derived from LCA data bases related to oil extraction from soybeans. Data published in these data bases were also used from the trans-esterification process. These basic data for oil extraction and trans-esterification starting wet algae were increased proportionally to the inlet flow rate [13].

4 Basic Inventory and Energy Balance

The main results of material and energy consumptions for 1 kg biodiesel production calculated by Lardon et al. [13] are shown in Table 2. This table shows that wet processes save energy needed for biomass drying, but need a higher microalgae production to get the same amount of biodiesel. This derives from the low efficiencies of downstream operations. Figure 2 shows also that normal cultures with biomass drying have a negative energy balance.

The cumulative energy demand in comparison with the products energy content estimated by Lardon et al. [13] for the four biodiesel production cases presented in Fig. 1 is shown in Fig. 3. In this figure, authors calculated the energy production on the basis of biodiesel and oilcake energy content, and energy demand by using the Ecoinvent database by assuming:

Table 2 Main material and energy consumption for biodiesel production from microalgae, basis 1 kg biodiesel [13]

	Normal		Low N	
	Dry	Wet	Dry	Wet
Algae culture and harvesting				
Algae (kg)	5.93	8.39	2.7	3.81
CO_2 (kg)	10.4	14.8	5.32	7.52
Electricity (MJ)	7.5	10.6	4	5.7
$CaNO_3$, as g N	273	386	29.4	41.6
Drying				
Heat (MJ)	81.8		37.1	
Electricity (MJ)	8.52		3.9	
Oil extraction				
Heat (MJ)	7.1	22.4	3.2	10.2
Electricity (MJ)	1.5	8.4	0.7	3.9
Hexane loss (g)	15.2	55	6.9	25
Oil trans-esterification				
Methanol (g)	114	114	114	114
Heat (MJ)	0.9	0.9	0.9	0.9
Total energy				
Consumption (MJ)	106.4	41.4	48.9	19.8
Production (MJ)	103.8	146.8	61	86
Balance (MJ)	−2.6	105	12	66

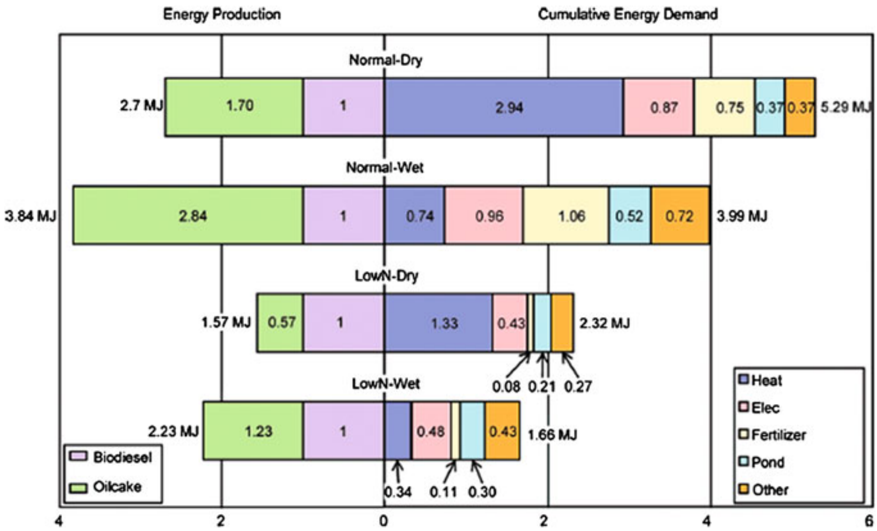

Fig. 2 Life cycle energy balance for biodiesel production from microalgae, basis 1 MJ biodiesel [13]

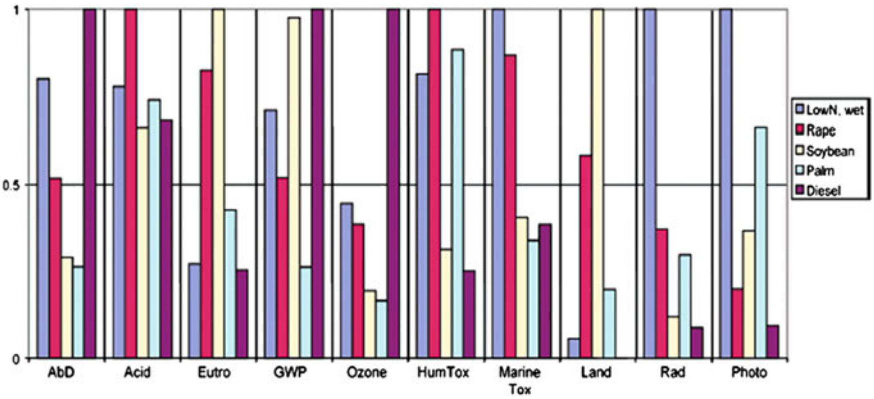

Fig. 3 Environmental impacts of biodiesel from microalgae (Low N wet) in comparison with commercial biodiesel and petroleum diesel depletion. Abiotic resources depletion (AbD), acid rain (Ac), water eutrophication (Eu), global warming potential (GWP), ozone layer depletion (Ozone), human toxicity (Hum Tox), marine toxicity (Marine Tox), land use (Land), ionizing radiations (Rad), photochemical oxidation (Photo)

- electricity produced with the European mix,
- burning natural gas to assure process heat,
- a 30 years life span for buildings, then their dismantling, recycling of steel and plastic material and concrete land filling,
- a 10 years basis replacement of electrical engines.

This figure clearly shows that only low nitrogen cultures with oil extraction from wet biomass present a positive energy balance. In this case, the larger energy demand is due to electricity (0.48 MJ/MJ biodiesel) followed by other operations, that is, facilities construction and dismantling (0.43 MJ/MJ biodiesel), heat requirements (0.34 MJ/MJ biodiesel), ponds operation (0.3 MJ/MJ biodiesel) and fertilizers production (0.11 MJ/MJ biodiesel).

Recently, Khoo et al. [20] have proposed different cultivation and oil extraction systems. These authors used:

- a *Nannochloropsis* strain with 25 % oil content in the base case and 35–45 % oil content under nitrogen limitation,
- a cultivation system consisting of photobioreactors for inocula preparation and open pond for large cultivation,
- a 25 gm^{-2} day^{-1} biomass productivity through the all year
- an harvesting system consisting of an air sparging assisted coagulation and flocculation apparatus capable to increase the biomass content from 0.1 % up to 3 % dry matter operating with 250 mg $FeCl_3 \cdot 6H_2O$/kg dry biomass as flocculants,
- a centrifuge for biomass dewatering up to 15 % dry matter,
- an extraction system based on biomass homogenization, and solvent extraction with an hexane-methanol 3:1 mixture used at 20:1 solvent-dry matter ratio.

According to experimental data [14–16], the assumptions appear realistic at laboratory scale, but concerns remain for large scale applications, particularly for centrifugation, homogenization and solvent removal from residual biomass. However, these authors found a cumulative energy demand ranging from 4.4 MJ/MJ biodiesel for the base case to 1.9 MJ/MJ biodiesel for the optimistic case (45 % oil content in the biomass, reduction of energy extraction from 2.5 to 1.5 MJ/MJ biodiesel and increase of the biodiesel heating value from 38 to 42 MJ/kg). Cultivation contributed on a minor measure to the total energy demands (0.56 MJ/MJ biodiesel) which are in line with those calculated by Lardon [13] and presented in Fig. 2.

Other authors report value rather different which depend on assumptions, climate and waste heat availability. For instance, Jorquera et al. [21] report energy demand ranging from 0.2 to 1.0 MJ/MJ biodiesel for plant located in Brazil not accounting for energy needed for oil extraction and trans-esterification. Clarens et al. [22] and Stephenson et al. [23] indicate 0.9–6.3 MJ/MJ biodiesel for plant located in the UK including anaerobic digestion units to get electricity from algal residues, and Baliga et al. [24] calculate from 0.44 to 0.67 MJ/MJ biodiesel for plants located in New York area, nearby a fossil fuel or biomass power plant capable to furnish waste heat for warming photo reactors cultivations inside greenhouse and for biomass drying.

5 Potential Environmental Impacts

Lardon et al. [13] calculated many impact categories for the four cases of biodiesel production presented above. Calculations have been performed by using the University of Leiden, Institute of Environmental Science (CML) methodology [25], and by allocating emissions between biodiesel and residual oil cake on the basis of their energy content. The following impact categories have been calculated: depletion of abiotic resources (AbD), acid rain (Ac), water eutrophication (Eu), 100 years global warming potential (GWP), 40 years ozone layer depletion (Ozone), 100 years human and marine toxicity (Hum Tox, Marine Tox), land competition (Land), emission of ionizing radiations (Rad) and photochemical oxidation (Photo). Details on the results as well to the contribution of single phases (energy production for algae cultivation and oil trans-esterification, consumptions for algae production excluding fertilizers, fertilizers, consumption in oil extraction and trans-esterification, combustion in internal combustion engines, wastewater treatment) are reported in Ref. [13]. Here, its worthy to point out that 90 % of energy consumption are needed for lipid extraction from dry biomass and 70 % from wet biomass. Accordingly, intensive research efforts should be dedicated to reduce energy consumption or use solar energy for drying or develop new technologies for wet lipid extraction.

In Fig. 3 results related to the best case (Low N wet) in comparison with the literature results of main commercial biodiesel and petroleum diesel are presented. These last data have been derived from Ecoinvent database and refer to biodiesel obtained from rapeseed in European scenarios, soybean in US scenario and palm oil imported in Europe from Malaysia.

Figure 3 show that biodiesel from microalgae has lower impacts in terms of water eutrophication and land use. This effect derives from a better use of fertilizers and the higher productivity of microalgae in comparison with oleaginous seeds. Contrarily, biodiesel from microalgae present a worst impact for categories related to the higher demand of fossil fuel (depletion of abiotic resources) and the higher use of fertilizers and electricity (depletion of the ozone layer, emission of ionizing radiation, photochemical oxidation). Accordingly, each options related to the reduction of these consumptions (new high lipid low protein strain, nutrient recycle also thorough anaerobic digestion of wastewater) should be deeply investigated.

Regarding global warming potential, the current crucial impact category, Fig. 3 shows that biodiesel from low nitrogen microalgae and wet extraction process has a higher impact in comparison with commercial biodiesel, except for soybean biodiesel. If data on GWP presented in Fig. 3 are seen in the context of the European directive on renewable energy (28/2009) which states 83.2 g CO_2 eq./MJ petroleum diesel, it can be estimated a GHG emission for the most favourable case of biodiesel production from microalgae (low nitrogen, wet extraction process) of 57.8 g CO_2 eq./MJ corresponding to 30.5 % GHG saving. This is almost in line with the current EU target (35 % GHG reduction) but well below for 2017 targets (50 % for existing plants and 65 % for new plants).

The strict comparison of environmental impacts reported by Lardon et al. [13] with those of other authors is even more difficult than energy demand because of the effect of different assumptions on the whole life cycle. However, Koo et al. [20], in agreement with Lardon et al. [13], clearly state that from the life cycle assessment perspective, biodiesel production does not necessarily reduce greenhouse gas emissions. Baliga et al. [24] state that in cold climates, the production of algae biodiesel with the utilization of waste heat rather than natural gas consumption is the only approach that reduces greenhouse gas emissions relative to soy biodiesel. Frank et al. [26], in a schematic based on the use of anaerobic digestion and methane burning in a combined heat and power system capable to generate power and heat for process purposes, highlighted the importance of considering the fate of the unrecovered nitrogen, specially that produces N_2O, a potent greenhouse gas with global warming potential 298 times that of CO_2.

6 Conclusions

Data collected and reviewed in this work indicate that among potential routes of CCU, biodiesel from microalgae is one of the most promising because of the direct utilization of solar radiation and the higher biomass and lipid areal productivity in comparison with marine or terrestrial plants. However, considering that:

- Solvent-based oil extraction technologies currently used for dry oily seeds (rapeseed, soybean, peanuts, etc.) can be used only on dry microalgae,
- fossil fuel demand for algae paste drying is much intensive,
- gravitational technologies presently used for oil extraction from wet oily seeds (olive and palm) cannot be used because of the nature of algae cells,

further research is still needed on oil extraction/trans-esterification from wet algae cells.

LCA studies on biodiesel production from microalgae published in the current literature are inhomogeneous, not only because of assumptions made for oil extraction and trans-esterification, but also because of a number of other assumptions such as for instance: water and nutrient recycle and the related fertilizer consumptions, use of residual biomass for feed purposes or for process heat and electricity supply through anaerobic digestion, use of grid-derived electricity from many primary energy mix, burning of different fossil fuel (natural gas or coal) for process heat supply, availability of waste heat from fossil or biomass power stations, climate of the cultivation area. Nevertheless, GHG savings appear in line with current standards required by 2009/28 directive (35 % lower than automotive-derived fossil fuel) only under the most favourable conditions, namely: biomass productivity at 25 $g.m^{-2}$ day^{-1} through the whole year, constant lipid content at 40–45 %, use of wet oil extraction processes still to be checked for large scale application but capable to keep total energy demand around 1.5 MJ/MJ biodiesel (value completely attributed to biodiesel and not allocated between biodiesel itself and residual biomass).

Under these conditions, biodiesel from microalgae shows lower impacts in comparison with rapeseed, soybean and palm oil biodiesel in terms of water eutrophication and land use, because of better use of fertilizers and the higher areal productivity of microalgae in comparison with oleaginous crops. Contrarily, biodiesel from microalgae presents a worst impact for categories related to the higher demand of fossil fuel (depletion of abiotic resources) and the higher use of fertilizers and electricity (depletion of the ozone layer, emission of ionizing radiation, photochemical oxidation). Accordingly, each options related to the reduction of these consumptions (new high lipid low protein strain, nutrient recycle also thorough anaerobic digestion of wastewater) should be deeply investigated.

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